

Carrier heating and high-order harmonics generation in doped graphene by a strong ac electric field

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The nonlinear response of electrons (holes) in doped graphene on ac pumping is considered theoretically for the frequency region above the energy relaxation rate but below the momentum and carrier-carrier scattering rates. Temporally-dependent heating of electrons by a strong ac field, which is described within the energy balance approach, leads to an effective generation of high-order harmonics. The efficiency of up-conversion of the 1 mm radiation into the third harmonic by a single-layer graphene is about 10^{-7} at pumping level ~ 100 kW/cm², room temperature, and concentration $\sim 5 \times 10^{11}$ cm⁻².

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The nonlinear response of carriers in graphene has been studied during the past years for the cases of the heating by a strong dc electric field and of the interband pumping in the visible or IR spectral regions. In the microwave frequency region, the nonlinear response is also effective because the massless and gapless energy spectrum and a suppressed relaxation of low-energy carriers. [1] It is important that different semiconductor structures is widely used for the multiplication (up-conversion) of the microwave pumping into THz radiation. [2] Recently, the frequency multiplier in the GHz region, which is based on the nonlinear response of a bipolar graphene near the electroneutrality point, have been demonstrated. [3] Such a multiplier should be effective in the frequency range $\omega \ll \nu_{gr}$ where ν_{gr} is the generation-recombination rate (in short devices, a fast contact injection can reduce this frequency restriction). For the high-frequency region, the collisionless regime of the high-order harmonics generation was considered in Ref. 4. These calculations are valid under the condition $\omega \gg \nu_m$ which is correspondent to the THz pumping because the momentum relaxation rate $\nu_m \sim 10^{13}$ s⁻¹ for a doped sample with the mean momentum \bar{p} . [1] The regime of nonlinear response under a strong pumping in the mm frequency range, when $\nu_m \gg \omega$, is not considered up to now.

In this paper we examine the heating of carriers and the high-order harmonics generation in doped graphene under a strong ac electric field $\mathbf{E}_t = \mathbf{E} \cos \omega t$ of the mm frequency range, so that $\omega \ll \nu_m$. We restrict ourselves by the weak anisotropy case, $eE/\nu_m \ll \bar{p}$, when the electron distribution function $f_{pt} + \Delta f_{pt}$ is written through the isotropic part, f_{pt} , and the frequency-independent anisotropic addition

$$\Delta f_{pt} \simeq \frac{e\mathbf{E}_t \cdot \mathbf{p}}{p\nu_p} \left(-\frac{\partial f_{pt}}{\partial p} \right), \quad (1)$$

where ν_p is the momentum relaxation rate. The symmetric part of distribution is governed by the averaged over

\mathbf{p} -plane equation (see similar calculations for heating by a dc field in Ref. 5)

$$\frac{\partial f_{pt}}{\partial t} + e\mathbf{E}_t \cdot \frac{\partial \Delta f_{pt}}{\partial \mathbf{p}} = \sum_r J_r(f_t|p). \quad (2)$$

Here the collision integrals $J_r(f_t|p)$ describe the nonelastic relaxation by acoustic phonons ($r = ac$) and the carrier-carrier scattering ($r = cc$). We neglect the relaxation by optical phonons because the energy of hot carriers is less than the optical phonon energy. The Joule heating contribution is described by

$$e\mathbf{E}_t \cdot \frac{\partial \Delta f_{pt}}{\partial \mathbf{p}} = \frac{(eE)^2}{2p} \frac{\partial}{\partial p} \left[\frac{p}{\nu_p} \left(-\frac{\partial f_{pt}}{\partial p} \right) \right] \cos^2 \omega t, \quad (3)$$

where a time dependency of f_{pt} appears due to the proportional to $\cos^2 \omega t$ factor.

The concentration of carriers and the current density, n and \mathbf{I}_t , are determined through f_{pt} and Δf_{pt} according to the standard formula

$$\left| \begin{matrix} n \\ \mathbf{I}_t \end{matrix} \right| = 4 \int \frac{d\mathbf{p}}{(2\pi\hbar)^2} \left| \begin{matrix} f_{pt} \\ e\mathbf{v}_p \Delta f_{pt} \end{matrix} \right|, \quad (4)$$

where $\mathbf{v}_p = v\mathbf{p}/p$ is the velocity of electron with the momentum \mathbf{p} and $v \simeq 10^8$ cm/s is the characteristic velocity of the linear dispersion law. Using Eq. (1) and introducing the nonlinear conductivity σ_t according to $\mathbf{I}_t = \sigma_t \mathbf{E}_t$, one obtains

$$\sigma_t = \frac{e^2 v}{\pi \hbar^2} \int_0^\infty \frac{dp}{\nu_p} \left(-\frac{\partial f_{pt}}{\partial p} \right) \approx \frac{e^2}{\pi \hbar^2} \frac{v}{v_d} f_t. \quad (5)$$

The right-hand part here is transformed for the case of the short-range scattering, when $\nu_p \simeq v_d p/\hbar$ is written through the characteristic velocity v_d ; this approach is valid up to energies ~ 100 meV. [6] Here $f_t \equiv f_{p=0t}$ stands for the time-dependent maximal distribution at $p = 0$.

Because of predominance of the intercarrier scattering in a heavily doped graphene if $\omega < \nu_{cc}$, the collision integral J_{cc} imposes the quasiequilibrium distribution $\tilde{f}_{pt} = f_t / [\exp(vp/T_t)(1 - f_t) + f_t]$ written through the

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time-dependent effective temperature, T_t , and f_t . These parameters are related by the normalization condition and the energy balance equation. Introducing the integrals over the dimensionless momentum

$$H_s(f_t) = \int_0^\infty \frac{dx x^s}{e^x(1-f_t) + f_t}, \quad s = 1, 2, \quad (6)$$

$$K(f_t) = \frac{1-f_t}{f_t} \int_0^\infty \frac{dx x^4 e^x}{[e^x(1-f_t) + f_t]^2},$$

one transforms the normalization requirement in Eq. (4) as follows

$$n = \frac{2}{\pi} \left(\frac{T_t}{\hbar v} \right)^2 f_t H_1(f_t). \quad (7)$$

The balance equation for the energy density, $4 \int d\mathbf{p} f_{pt} v p / (2\pi\hbar)^2$, is transformed into

$$\begin{aligned} & \frac{\partial}{\partial t} \left[T_t \frac{2}{\pi} \left(\frac{T_t}{\hbar v} \right)^2 f_t H_2(f_t) \right] - \sigma_t E_t^2 \\ &= \frac{v_e}{v} \frac{2}{\pi} \left(\frac{T_t}{\hbar v} \right)^2 \frac{T_t^2}{\hbar} \left(1 - \frac{T_t}{T_{ph}} \right) K(f_t), \end{aligned} \quad (8)$$

where the Joule contribution was written through the time-dependent conductivity $\sigma_t \propto f_t$, see Eq. (5). The energy relaxation contribution in the right-hand side of (8) is written for the case of the quasielastic scattering by acoustic phonons at temperature T_{ph} . [5] Here we used the energy relaxation rate $v_e p / \hbar$ written through the characteristic velocity $v_e \ll v, v_d$.

Numerical solution of Eq. (8) is performed under the relation (7) and the periodicity conditions $T_{t+\pi/\omega} = T_t$ and $f_{t+\pi/\omega} = f_t$. The nonequilibrium distribution \tilde{f}_{pt} is determined through $T_t = T + \delta T_t$ and $f_t = f + \delta f_t$. Here we separated the time-averaged parts, T and f , which describe hot electrons, and the oscillating contributions, δT_t and δf_t . The calculations are performed at $T_{ph} = 300$ K for the concentrations $n = 5 \times 10^{11} \text{ cm}^{-2}$ and 10^{12} cm^{-2} , the intensities up to 200 kW/cm^2 , and the characteristic velocities $v_d \simeq 3 \times 10^7 \text{ cm/s}$ and $v_e \simeq 51.4 \text{ cm/s}$. The dependencies of time-averaged contributions T and f on the pumping intensity S are shown in Figs. 1a and 1b. At low intensities of pumping $S < 5 \text{ kW/cm}^2$, T_t and f_t vary strongly in analogy with to the results on heating by a dc electric field. [5] A slowness of these dependencies takes place at higher pumping levels, in the region $S \sim 10 - 100 \text{ kW/cm}^2$. Since the relaxation rates increase with energy, both T_t and f_t increase with concentration. Notice, that a response on a probe dc field (photoconductivity) is described by Eq. (5) with the time-averaged f so that Fig. 1b determines the photoconductivity versus S .

The time-dependent contributions $\delta T_t = T_t - T_{t=0}$ and $\delta f_t = f_t - f_{t=0}$ are shown in Figs. 2a and 2b for the region $S = 25 - 100 \text{ kW/cm}^2$ at concentration $5 \times 10^{11} \text{ cm}^{-2}$.

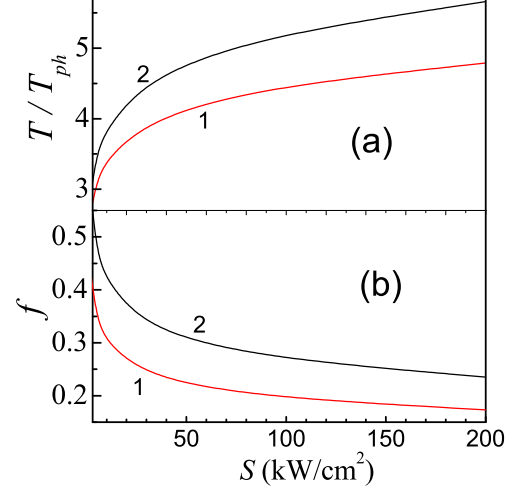


FIG. 1: Time-averaged temperature T (a) and maximal distribution f (b) versus pump intensity S plotted for concentrations $5 \times 10^{11} \text{ cm}^{-2}$ (1) and 10^{12} cm^{-2} (2).

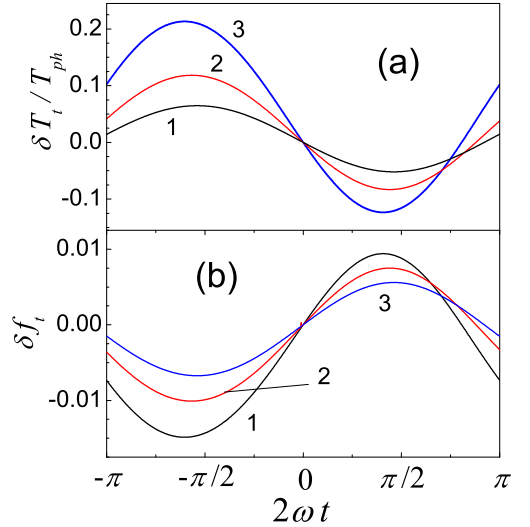


FIG. 2: Nonharmonic time-dependent contributions to temperature δT_t (a) and to maximal distribution δf_t (b) for the concentration $5 \times 10^{11} \text{ cm}^{-2}$ and the 1 mm pumping intensities 25 kW/cm^2 (1), 50 kW/cm^2 (2), and 100 kW/cm^2 (3).

In particular, δf_t determines the time-dependent part of σ_t , i. e. the nonlinear current under a strong pumping. Similar dependencies for the concentration 10^{12} cm^{-2} are not shown.

Further, we discuss the efficiency of transformation of the millimeter pumping into THz signal for the case of normal propagation both the pumping wave and the k th order response determined by the field $\mathbf{E}_z \exp(-ik\omega t)$. Substituting \tilde{f}_{pt} into the 2D current density of Eq. (4) one obtains the Fourier expansion $\mathbf{I}_t = \sum_k \mathbf{I}_{k\omega} \exp(-ik\omega t)$ with the nonzero odd harmonics (it is due to in-plane isotropy of the problem). The

k th harmonic of in-plane THz field, \mathbf{E}_z , is governed by the wave equation: [7]

$$\frac{d^2 \mathbf{E}_z}{dz^2} + \epsilon \left(\frac{k\omega}{c} \right)^2 \mathbf{E}_z + i \frac{4\pi}{c^2} k\omega \mathbf{I}_{k\omega} \delta_N(z) = 0, \quad (9)$$

where ϵ is the uniform dielectric permittivity and $\delta_N(z)$ is the transverse form-factor of the nonlinear current calculated above. For the N -layer graphene stack, this factor is normalized as $\int_{-d/2}^{d/2} dz \delta_N(z) = N$ with $d \rightarrow 0$. The solution of Eq. (9) at $z \neq 0$ takes form $\mathbf{E}_z = \mathbf{E}_{k\omega} \exp(i\kappa_{k\omega}|z|)$ with the wave vector $\kappa_{k\omega} = \sqrt{\epsilon}k\omega/c$. The amplitude of k th harmonic, $\mathbf{E}_{k\omega} = -N\mathbf{I}_{k\omega}(2\pi/\sqrt{\epsilon}c)$ is obtained from the boundary condition at $z \rightarrow 0$.

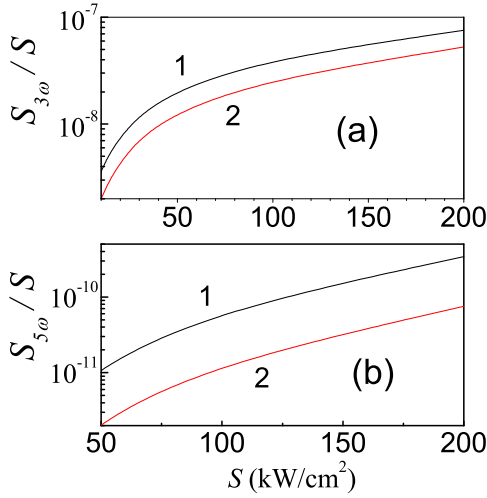


FIG. 3: Efficiency of transformation into k th harmonics, $S_{k\omega}/S$, versus pumping intensity S for $k=3$ (a) and $k=5$ (b) at doping levels $5 \times 10^{11} \text{ cm}^{-2}$ (1) and 10^{12} cm^{-2} (2).

The efficiency of up-conversion of 1 mm pumping of intensity S into THz signal of intensity $S_{k\omega}$ is determined by the ratio $S_{k\omega}/S = 4|\mathbf{E}_{k\omega}|^2/E^2$. Substituting $\mathbf{I}_{k\omega}$ from Eq. (4) we plot this efficiency for $k=3$ and 5, see Figs. 3a and 3b, where third and fifth harmonics are correspondent to 0.9 THz and 1.5 THz signals, respectively. One

obtains the output intensities $S_{3\omega} \sim 1.1 - 8.9 \text{ mW}/\text{cm}^2$ for the pumping range 50 - 150 W/cm^2 at concentration $5 \times 10^{11} \text{ cm}^{-2}$ while $S_{5\omega}$ corresponds up to $\sim 10 \text{ } \mu\text{W}/\text{cm}^2$ range. Since $S_{k\omega} \propto N^2$ for the N th layer structure of epitaxial graphene, [8] the up-converted 0.9 THz signal appears to be about 0.1 W/cm^2 (or $\sim 0.2 \text{ mW}/\text{cm}^2$ at 1.5 THz) under the 100 W/cm^2 pumping at the $5 \times 10^{11} \text{ cm}^{-2}$ doping level and $N=5$.

Next we briefly discuss the assumptions used in our calculations. We describe the heating of carriers with the use of the quasiequilibrium distribution approach. This is valid for the heavily doped graphene which is suitable for an efficient up-conversion. Typically, $\nu_m \sim \nu_{cc}$ in doped samples and, together with the low-frequency condition $\omega < \nu_m$, we arrive at the restriction $\nu_e < \omega < \nu_m$, ν_{cc} which is correspondent to the $10^9 \text{ s}^{-1} - 10^{13} \text{ s}^{-1}$ frequency region. In spite of $f_{p=0t} < 1$ we neglected the interband generation-recombination processes because the density of states vanishes at low energies. Also, the interband optical transitions are not essential in the spectral region up to 5ω . Beside of this, we perform the simplified electro-dynamical estimates of the efficiency of up-conversion for the case of the normal propagation of radiation through the structure with the same dielectric permittivities of the substrate and cover layers. The simplifications listed do not change either the character of the high-order nonlinear response or the numerical estimates for the efficiency of up-conversion.

In summary, we have demonstrated that the heating by a strong ac field results in an essentially nonharmonic response. This leads to the effective generation of the third harmonic and opens a possibility for the efficient up-conversion of mm pumping into THz signal. An experimental study of the process considered and the electro-dynamical calculations of a graphene structure integrated to an antenna or into a THz resonator are very opportunely now.

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